

# Bulk and local properties of intermetallic GdAgSn compound

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**Abstract.** The results of magnetic susceptibility as well as electric resistivity measurements complemented with <sup>155</sup>Gd Mössbauer spectroscopy investigations carried out within wide temperature range for GdAgSn compound are discussed.

## 1. Introduction

The ternary equiatomic stannides (R, A)TSn (R or A = rare earth or actinide element, respectively; T = 3d, 4d or 5d transition element) have intensively been studied with respect to their largely varying magnetic and electrical properties [1-3]. Among them the title compound reveals interesting magnetic behaviour as it was shown by the AC susceptibility and Mössbauer measurements at low temperatures [4, 5]. It has been found that on cooling down this compound orders first antiferromagnetically at  $T_N=34.3(2)$  K, while at  $T_f = 15.3$  K undergoes spin glass-like transition (SG).

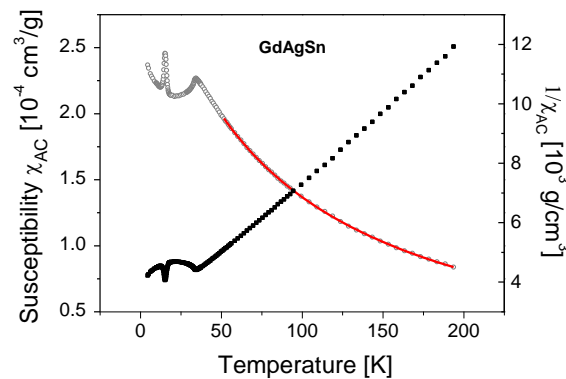
## 2. Experimental

Our polycrystalline GdAgSn sample was synthesized by arc melting the constituent elements with purity better than 99.9% in argon atmosphere and consecutive annealing procedure for better homogeneity as previously described [4]. The X-ray powder diffraction pattern was recorded at room temperature with a Siemens D-501 diffractometer using the Ni-filtered  $\text{CuK}_\alpha$  and the results has already been published elsewhere [4, 5]. Magnetic susceptibility and magnetization measurements were carried out with a 7225 Lake Shore AC Susceptometer/DC Magnetometer operating in AC mode on powdered samples in the temperature range 4.2 K – 200 K and in the external magnetic fields up to 60 kOe. The electrical resistivity ( $\rho$ ) measurements were performed by means of Physical Property Measurement System (Quantum Design, PPMS-9T) on a bulk probe using a steady-current, standard four point technique and applying an excitation current of 80 mA in zero external magnetic field. Our specimen had of approximate dimensions 1x1x5 mm<sup>3</sup>. (86.5 keV ( $I_g = 3/2$ ,  $E1$ ,  $I_e = 5/2$  transition)) <sup>155</sup>Gd Mössbauer spectra were collected in a transmission geometry cryostat using a conventional constant-acceleration spectrometer of the Kankeleit type. The 20 mCi <sup>155</sup>Eu:SmPd<sub>3</sub> source was kept at 4.2 K to increase the efficiency of the resonance emission. The temperature of the absorber was varied between 4.2 and 50 K with a stabilisation better than 0.05 K. In contrast to the fitting method applied in our previous investigations (where Lorentzian approximation was used [5]), for the proper description of the recorded <sup>155</sup>Gd Mössbauer spectra a least-squares fitting procedure which includes the diagonalization of the full hyperfine Hamiltonian within the transmission integral approximation was used.



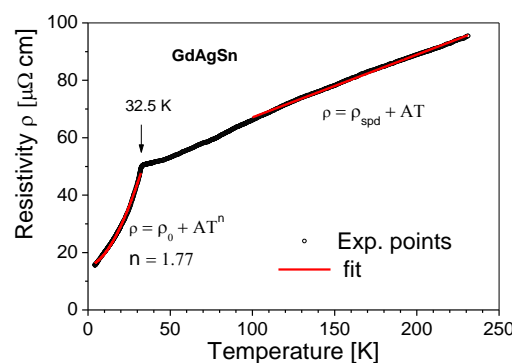
### 3. Results and discussion

X-ray diffraction investigations made on the polycrystalline GdAgSn sample revealed the existence of only single phase and confirmed that this compound crystallizes in the hexagonal  $\text{CaIn}_2$ -type of structure with space group  $P6_3/mmc$  and the derived lattice parameters are following:  $a = 470.9(1)$  pm and  $c = 742.5(3)$  pm [2]. The temperature dependence for the absolute value of the magnetic AC susceptibility  $\chi_{AC} = [(\chi')^2 + (\chi'')^2]^{1/2}$  and its reciprocal are displayed in figure 1.



**Figure 1.** Temperature dependence for the absolute value of the magnetic ac susceptibility  $\chi_{AC} = [(\chi')^2 + (\chi'')^2]^{1/2}$  (left-hand scale) and its reciprocal (right-hand scale) recorded for GdAgSn in zero external magnetic field at the internal frequency  $f = 375$  Hz and the applied oscillating field strength  $H_{AC} = 5$  Oe. The solid line is a result of the fitting procedure in the paramagnetic range, for details see the text.

The temperatures observed here for both maxima are in perfect agreement with those previously published [4, 5] i.e. a phase transition from a paramagnetic to an antiferromagnetic state (PM→AFM) takes place at the Néel temperature  $T_N = 34.3(2)$  K while the second one at  $T_i = 15.3(2)$  K is that which have been ascribed to the spin-glass transition. Above 52 K (see Fig. 1), the recorded susceptibility obeys fairly well a modified Curie-Weiss law in the form:  $\chi_{AC} = \chi_0 + C/(T - \theta_p)$  with the temperature independent factor  $\chi_0 = -1.21 \cdot 10^{-6}$  cm<sup>3</sup>/g, the Curie constant  $C = 2.092 \cdot 10^{-2}$  Kcm<sup>3</sup>/g, and the paramagnetic Curie temperature  $\theta_p = -53.7$  K. The negative value of  $\theta_p$  is indicative for antiferromagnetic correlations. The effective magnetic moment was derived from the formula  $\mu_{eff} = 2.83(MC)^{1/2}$  where  $M$  is the molar mass. The experimental value  $\mu_{eff} = 8.01 \mu_B$  is somewhat higher but agrees well with the theoretical Russel-Saunders value  $\mu_{eff} = g\mu_B[J(J+1)]^{1/2} = 7.94 \mu_B$  for the free  $\text{Gd}^{3+}$  ion. Figure 2 shows the temperature variation of electrical resistivity recorded at zero external magnetic field.



**Figure 2.** Temperature dependence of electrical resistivity recorded for GdAgSn. The continuous lines present the least-squares fits to the low and high temperature parts of  $\rho(T)$  according to the given formulas below each part, respectively.

The linear behavior at high temperatures and the positive temperature coefficient of resistivity ( $d\rho/dT > 0$ ) indicate the metallic nature of the compound. The striking feature is a lack of any resistivity anomaly around  $T_f = 15.3$  K while a drastic change of resistivity slope is registered at  $T = 32.5$  K, i.e. close to  $T_N = 34.3$ . It has to be underlined here, that such an observation is in line with the proposed spin glass-like character of GdAgSn below  $T_f$  and its ordered antiferromagnetic character below  $T_N$  [4,5]. As a matter of fact, the transport properties do not reveal any transition at spin glass temperature but only a gradual transformation which is associated with the lack of ordered state (see [6]). Below  $T_N$  we have found that the best fit to the experimental data in an expanded range (see Fig. 2.) may be approximated by the simple power law in the form:  $\rho(T) = \rho_0 + AT^n$  with the  $\rho_0 = 15.44 \mu\Omega\text{cm}$ ,  $A = 0.069 \mu\Omega\text{cm/K}^{1.77}$  and  $n = 1.77$ . At high temperatures ( $T > T_N$ ) the resistivity  $\rho_{\text{mag}}(T) \equiv \rho_{\text{spd}}(T) = \text{constant}$ , where  $\rho_{\text{spd}}$  is the spin-disorder contribution and the phonon contribution  $\rho_{\text{ph}}(T)$  is fairly well linear function of  $T$  [7]. Thus, in this temperature region the formula:  $\rho(T) = \rho_{\text{spd}} + AT$  can be fitted. The estimated in such a way values of  $\rho_{\text{spd}}$  and  $A$  are  $45.27 \mu\Omega\text{cm}$ , and  $0.218 \mu\Omega\text{cm/K}$ , respectively. Low temperature  $^{155}\text{Gd}$  Mössbauer spectra obtained around and below  $T_N$  for GdAgSn can be fitted with the single magnetic hyperfine component rather well, and no drastic change can be registered in the shape of these spectra below  $T_f$  in spite of expected field distribution characteristic for spin glass state. The obtained quadrupole interaction constant  $\Delta E_Q = eQ_s V_{zz} = -1.592(2)$  mm/s can be easily converted into value of the electric field gradient (EFG)  $V_{zz}$  at the gadolinium nuclei by the formula:  $V_{zz} = 2.2206 \cdot 10^{21} \Delta E_Q [\text{mm/s}] \text{ V/m}^2 = -3.535(4) \times 10^{21} \text{ V/m}^2$ . Furthermore, knowledge of the EFG at the Gd site allows an estimation of the quadrupolar term  $B_2^0$  in the Stevens expansion of the crystal field Hamiltonian:  $\sum B_n^m O_n^m$ . This term is directly related to  $\Delta E_Q(^{155}\text{Gd})$  by the expression:

$$B_2^0 [\text{K}] = -\alpha * \langle r^2 \rangle_{4f} * 90.2 * \Delta E_Q (^{155}\text{Gd})$$

Here,  $\alpha$  is the appropriate Stevens factor, the mean squared radius  $\langle r^2 \rangle_{4f}$  is expressed in atomic units and  $\Delta E_Q$  in mm/s. Under assumption that  $B_2^0$  is the leading term in the crystal field Hamiltonian, then when translated for other rare earth ions in the family of isostructural RAgSn compounds, where the rare earths metals have non-zero angular momentums its sign gives information about the magnetocrystalline anisotropy, since then  $H_2^0 (\text{CEF}) = B_2^0 [3J_z^2 - J(J+1)]$ . For a negative  $B_2^0$  the ground state doublet is built up of states with a maximum absolute value of  $J_z$  which means that the magnetic moment is pointed in the direction of the  $z$ -quantization axis. In contrary, for positive values of  $B_2^0$  only states with the possible smallest  $|J_z|$  contribute to the electronic ground state and the magnetic moment is aligned in the basal plane ( $\perp z$ ).

## Conclusions

All carried measurements confirm very well the transition observed at  $T_N$ , however, the transition at  $T_f$  has no noticeably influence on the shape of  $^{155}\text{Gd}$  spectra what could be explained by an assumption that the local nearest-neighbour  $\text{Gd}^{3+}$  spin configuration below  $T_f$  is not significantly modified.

## Acknowledgments

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